Air/Sea Transfer of Gases and Aerosols

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LONG-TERM GOALS

Our long range goals for this project are to: 1) understand the effect of various physical and chemical properties of the air-sea interface on gas exchange, and 2) characterize the sea surface texture and turbulent boundary layers. These studies will provide a basis for future work on the active and passive microwave remote sensing of the sea surface and the estimation of surface wind vectors and gas transfer velocities from such information. They will provide new insight into the mechanisms by which chemicals are transferred across the air/sea interface.

OBJECTIVES

This project is a collaborative experimental study of wave dynamics, boundary layer turbulence, and gas exchange in a salt-water wind-wave tank. Our objectives are: 1) to relate surface texture and boundary layer turbulence to imposed surface wind stress and gustiness, and atmospheric stability to provide insight into the factors controlling remote sensing of the ocean surface, and 2) to relate direct measurements of air-sea gas fluxes to the surface water chemistry and texture and boundary layer turbulence. To our knowledge this is the first attempt to use direct flux measurements to study gas exchange in the laboratory.

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14. ABSTRACT
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APPROACH

Our approach is to carry out laboratory experiments under controlled conditions, in which the state of the fluids and interface can be well characterized. A new chemical ionization mass spectrometer (referred to as CIMS) was designed and built to make fast response chemical measurements for these studies. The experiments are being carried out using a new facility, the Air-Sea Interaction Salt-water Tank (ASIST) that has recently been constructed at RSMAS, University of Miami. ASIST is a linear, recirculating wind/wave tank. During these experiments, water surface textures are characterized using an imaging slope gauge and scanning slope gauge. Turbulence measurements in air and water are made using hot x-films and a conical hot film probe carried by a wave follower. Turbulence measurements and visualization in the water are also made using particle image velocimetry. Gas exchange is studied using eddy correlation flux measurements involving fast-response chemical detection for carbon dioxide via IR absorption, and dimethylsulfide (DMS) by CIMS. The key personnel involved in the project are Drs. Eric Saltzman (gas exchange), Mark Donelan (turbulence, wave properties, and remote sensing), and Warren De Bruyn (mass spectrometry).

WORK COMPLETED

Most of the work in this project has focused on constructing and testing the various components of the experiment. Preliminary flux experiments have been carried out in the wind/wave tank to assess the performance of various systems. The construction of a fast response chemical ionization mass spectrometer has been completed, and it has been optimized for detection of DMS in terms of chemistry and source optics. A wind/wave tank experiment combining physical and chemical measurements was conducted during August, 2002. The full analysis of the data resulting from the experiment has not yet been completed, but initial results suggest that the scientific goal of linking observations of water turbulence to direct flux measurements will be achieved. This report contains a summary of preliminary results from this experiment.

RESULTS

Direct gas flux measurements were made in the wind/wave tank using both eddy correlation and profile techniques. These experiments provided the first critical test of the CIMS instrument's capability to make fast-response chemical measurements under marine conditions. For these experiments, the tank was configured in open flow mode, with a single pass of outdoor air over the water surface. The water concentration of DMS and CO_2 were maintained at levels of approximately 1 and 10^4 ppm, respectively by injection of the pure compounds. Figure 1 is a plot illustrating the frequency response of the CIMS for DMS. Under the conditions of this experiment, DMS measurements were acquired by the CIMS at 20 Hz. The actual frequency response was 8 Hz, as a result of diffusion in the inlet tubing and data binning. An example of the instrument response is shown in Figure 1.

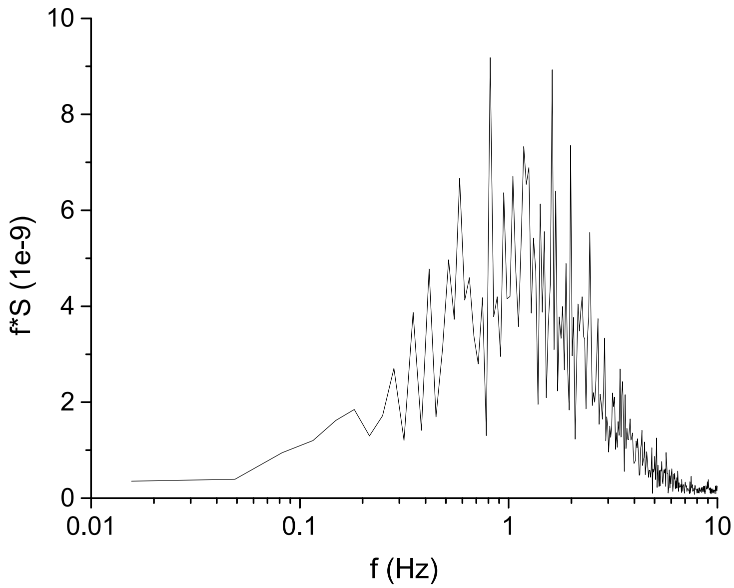

Figure 1. Frequency response (frequency-weighted) of CIMS instrument to DMS in the wind/wave tank, for a wind speed of approximately 14 m/s.

On the small turbulence scales of the wind/wave tank, a frequency response of 8 Hz does not capture the entire gas flux, particularly at high wind speeds. Fast response temperature measurements (100 Hz) allowed the entire range of turbulent scales to be captured. For example, Figure 2 is a plot of the cumulative cospectra of temperature and the vertical wind velocity component, collected for wind speeds ranging from 1 to 10 m/s. By plotting the cumulative cospectrum of vertical winds and temperature (normalized by height and wind speed), a "universal curve" is developed, which can be used to correct the chemical fluxes to account for the total gas flux. Figure 3 shows the same data sets transformed onto universal coordinates. Note how the variability from data collected at different heights and wind speeds collapses into a narrow range. Figure 4 illustrates the cospectra of DMS and the vertical component of the winds, also plotted in univerals coordinates.

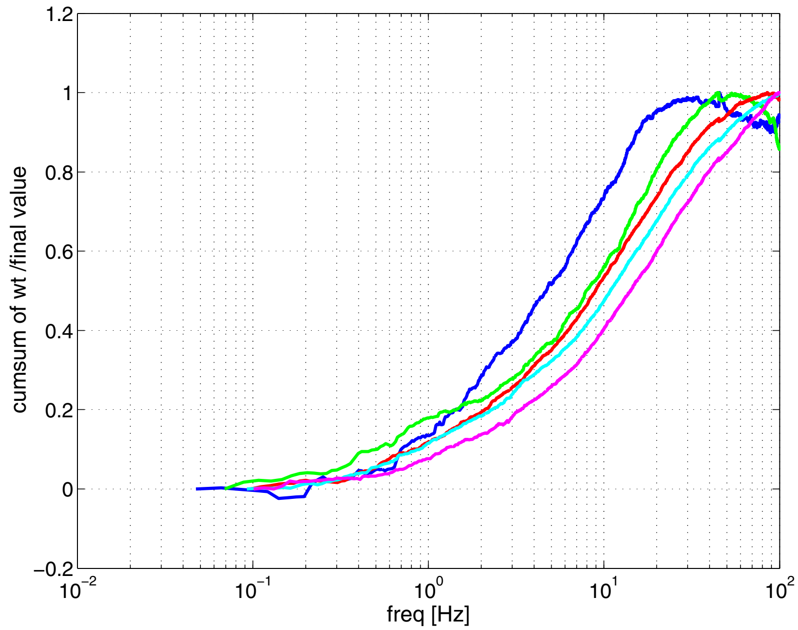

Figure 2. Cumulative sum of cospectra of temperature and vertical winds for wind speeds ranging from 1 to 10 m/s.

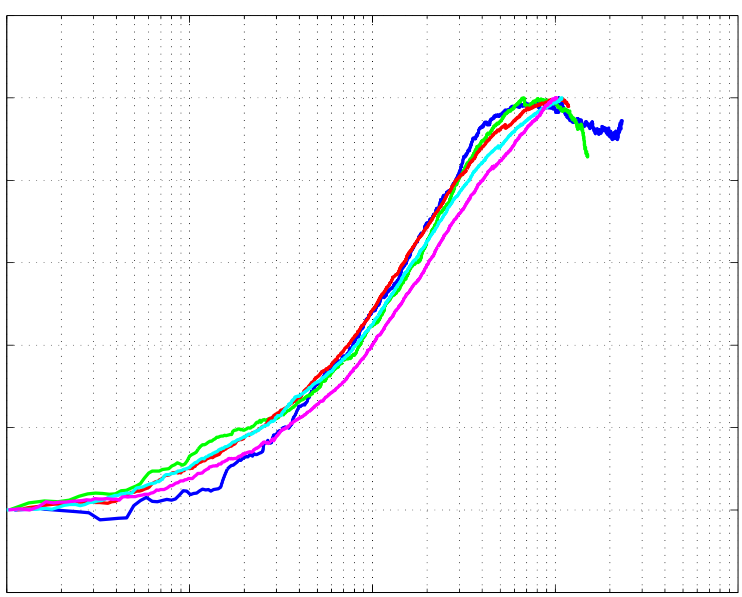

Figure 3. Cumulative sum of cospectrum of vertical wind speed and temperature (shown in Figure 2 above) plotted in universal coordinates.

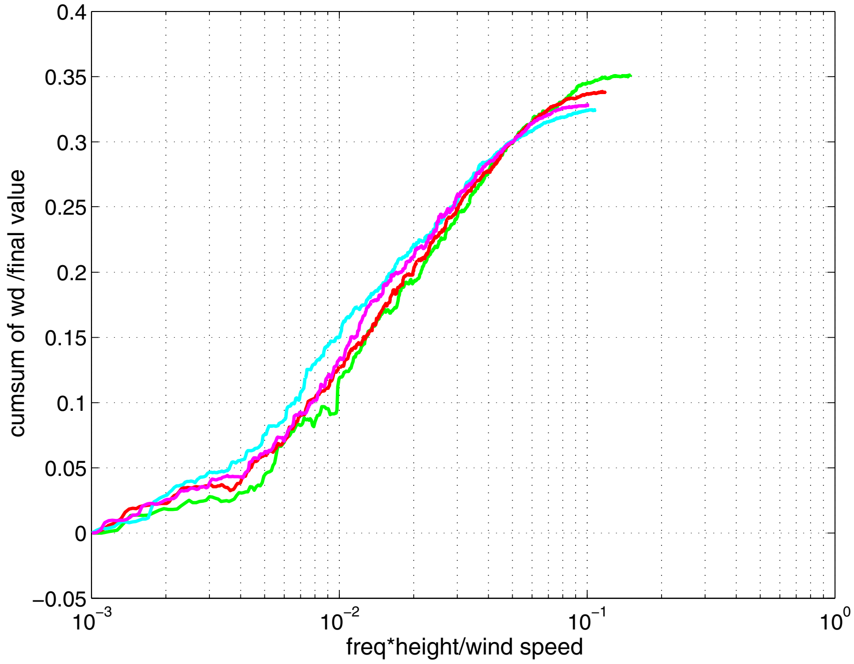

Figure 4. Cumulative sum of cospectrum of DMS and vertical wind speed, plotted in universal coordinates.

Profile measurements were also made during the experiment, by varying the height of the gas inlet above the water surface (Figure 5). Such profiles are not possible for CO_2, when the wind/wave tank is in open mode. In this configuration, fresh air is continually drawn in from outside and external variations in CO_2 dominate the mean level of CO_2 in the tank. Of all parameters measured, DMS is least influenced by external conditions. Although the analysis of this data set is ongoing, preliminary analysis suggests reasonable agreement between DMS fluxes measured by the eddy correlation and profile techniques.

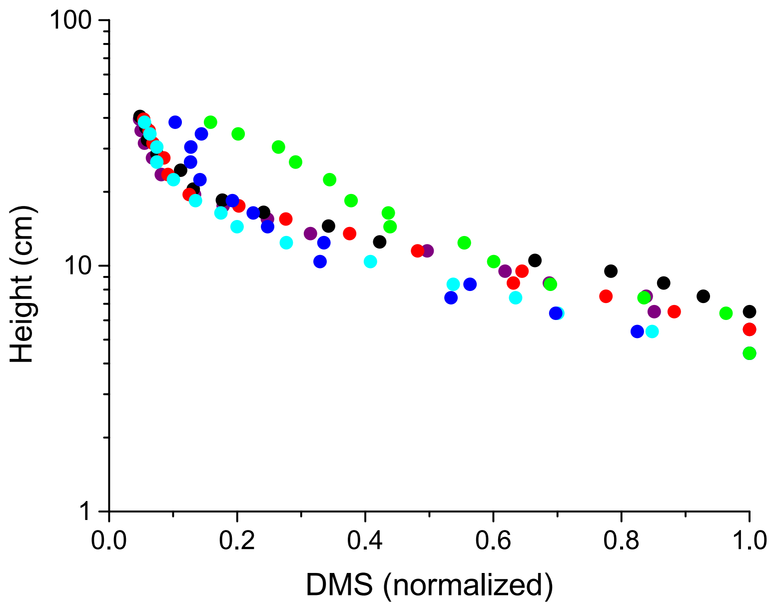

Figure 5. Vertical profiles of DMS in the wind/wave tank ranging at wind speeds ranging from 2.5-15 m/s. The DMS levels are normalized to the concentration at the lowest height. The slope of the lowermost 5 points are used to estimate the DMS flux.

Measurements of the DMS partial pressure in the water were made before and after each gas flux measurement. From the DMS flux and water-air concentration gradient, we can calculate the gas exchange coefficient or piston velocity (k). Preliminary analysis of the profile data yields the k vs wind speed relationship shown below in Figure 6. The data shown are in arbitrary units, as final scaling of the data is ongoing. Full analysis of the flux data set will provide similar relationships for both DMS and CO_2 from eddy correlation measurements, along with wave height, wave slope, and water turbulence measurements which were collected simultaneously with the gas fluxes. This is a unique data set that demonstrates the first successful use of direct flux measurements in laboratory gas exchange studies. The versatility of the CIMS instrument will allow future experiments to involve a wider range of chemical species, covering a broader range of Schmidt numbers and solubilities.

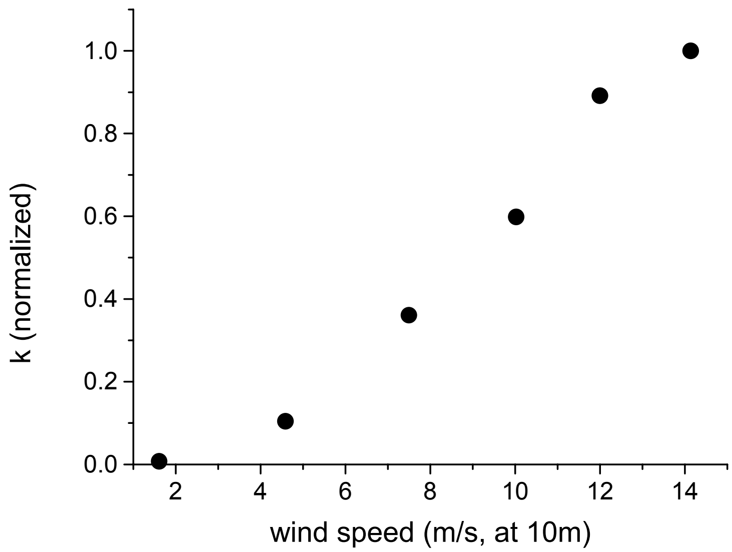

Figure 6. Preliminary data illustrating the relationship between DMS gas exchange coefficient and wind speed (at 10m). The data are based on fluxes derived from profile measurements in the wind/wave tank.

TRANSITIONS

We expect that this project will eventually result in two types of transitional developments: 1) a flux measurement capability for eddy-correlation measurements of trace gases at sea, and 2) improved algorithms relating the state of the air/sea interface to remotely sensed properties. For the CIMS instrument, the transition from measurement of fluxes in the wind/wave tank to the sea surface appears feasible. The laboratory studies and modeling of instrument response characteristics suggest that the CIMS instrument is capable of making accurate flux measurements under real-world field conditions. A field test will be carried out during the next few months.

RELATED PROJECTS

This project is closely related to a DURIP award for the construction of the ASIST wind/wave facility.